

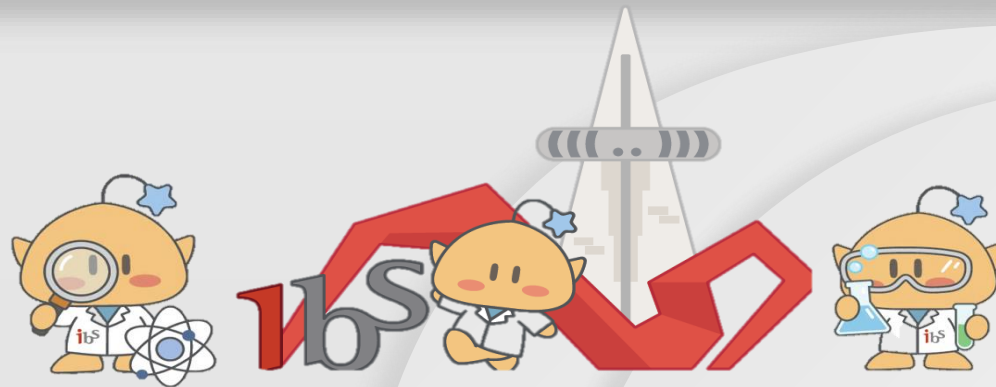
LOW
RADIOACTIVITY
TECHNIQUES

2026
WORKSHOP X

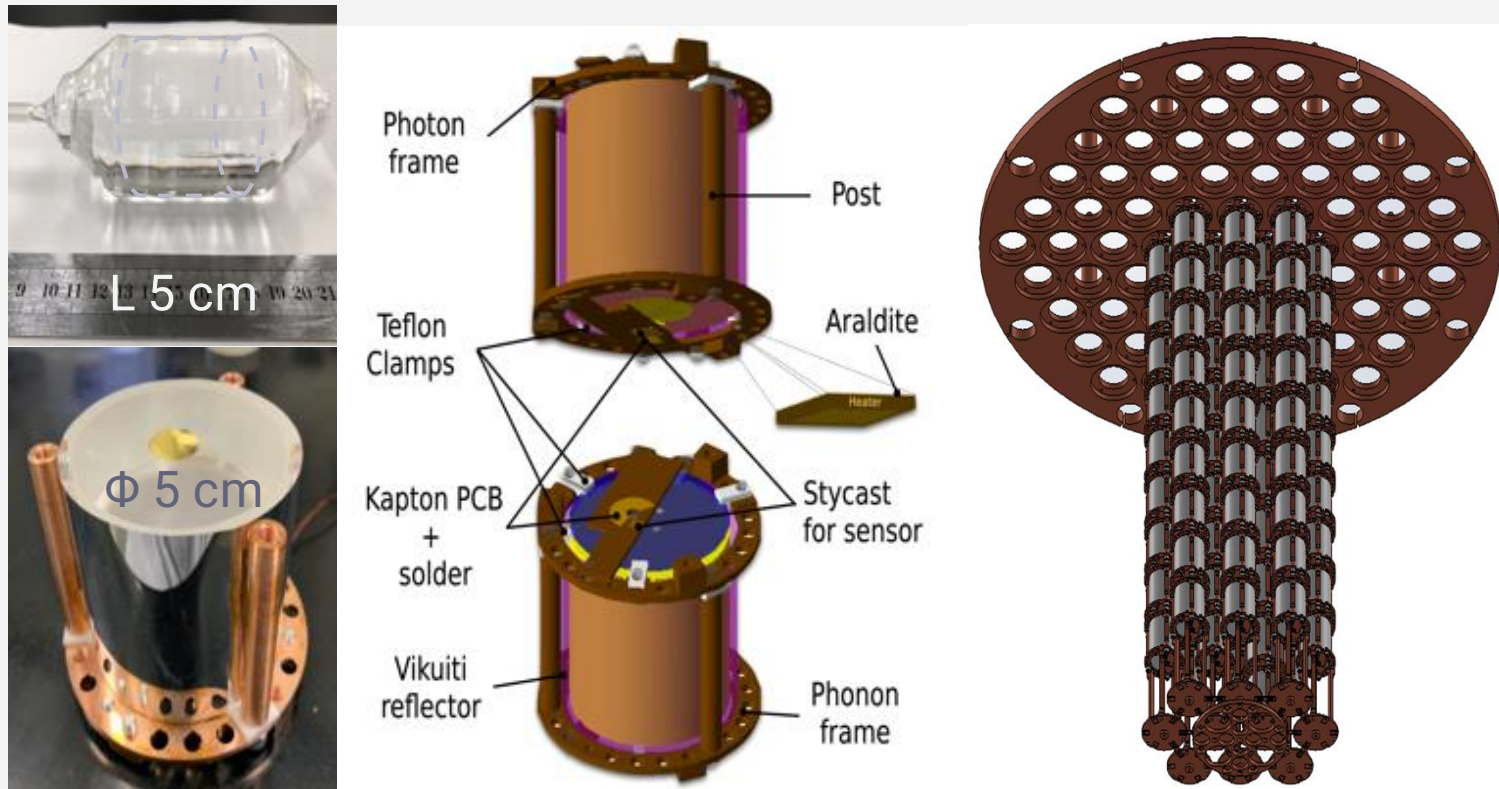


Material procurement, purification, and assay for low-background experiments at CUP

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24 September 2026



AMoRE-II and 360 $\text{Li}_2^{100}\text{MoO}_4$ crystals are searching for $0\nu\beta\beta$



- AMoRE searches for the neutrinoless double beta decay ($0\nu\beta\beta$) of ^{100}Mo using 360 bolometric $\text{Li}_2^{100}\text{MoO}_4$ crystals.
- Background index of $\sim 10^{-4}$ c/kg with a mass of the studied isotope Mo-100 of interest of 100 kg.
- Target intrinsic purity for Th and U in the crystals is to be below $10 \mu\text{Bq/kg}$

I. CUP-grown $\text{Li}_2^{100}\text{MoO}_4$ crystal ingot; II. $\text{Li}_2^{100}\text{MoO}_4$ scintillation element after cutting, polishing, surface cleaning, and gold film deposition procedures; III. Copper crystal holder frame and near-crystal components; IV. Detector tower consisting of 90 enriched $\text{Li}_2^{100}\text{MoO}_4$ crystals for the first stage of AMoRE-II.

Yemi Underground Lab

houses AMoRE-II experiment



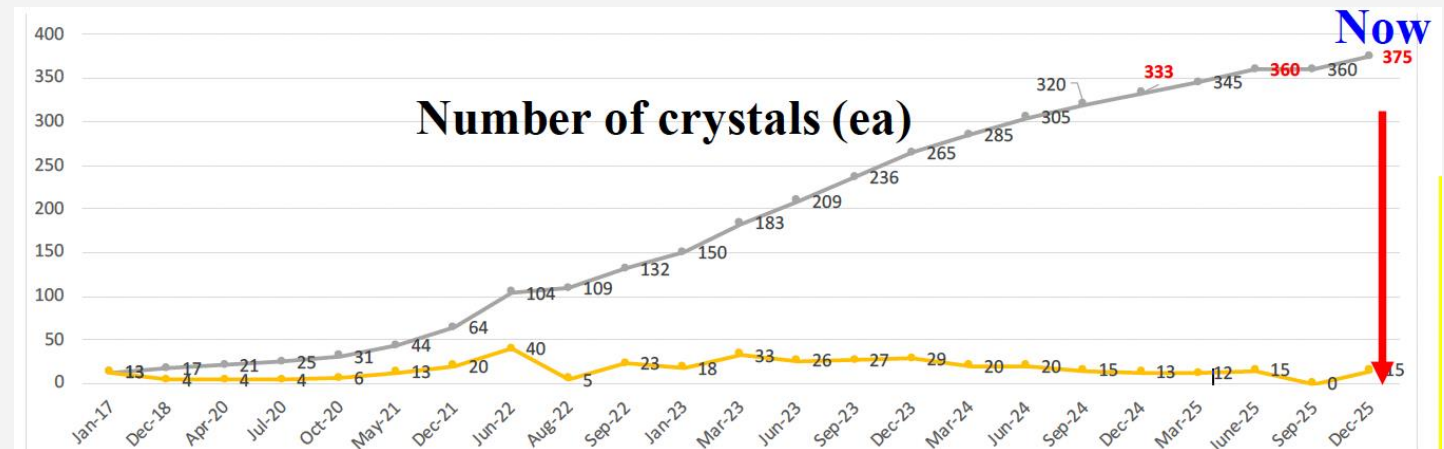
- Total area : 10,000 m² including 3,000 m² Lab. space.
- Overburden : 1009 m from the top of the Mt. Yemi. (-120m)

LMO crystal production history

- From 2016 to 2019, we received **120 kg of Mo-100 isotope**, which is equivalent to about 180 kg of $^{100}\text{MoO}_3$
- Three years of R&D** let us develop a method of purification for $^{100}\text{MoO}_3$ and its recycling and re-purification from the melt that remains after the crystal growth, and another **two years to scale up** and adjust the facility for a stable production rate of 5 kg per month of purified powder
- We recycle all the by-products:** LMO melts, purification by-products, cutting and polishing leftovers and tissues, etc.

TABLE 1 Range of concentrations for measured contaminants in different lots of raw $^{100}\text{MoO}_3$ powder. * Data of the producer.

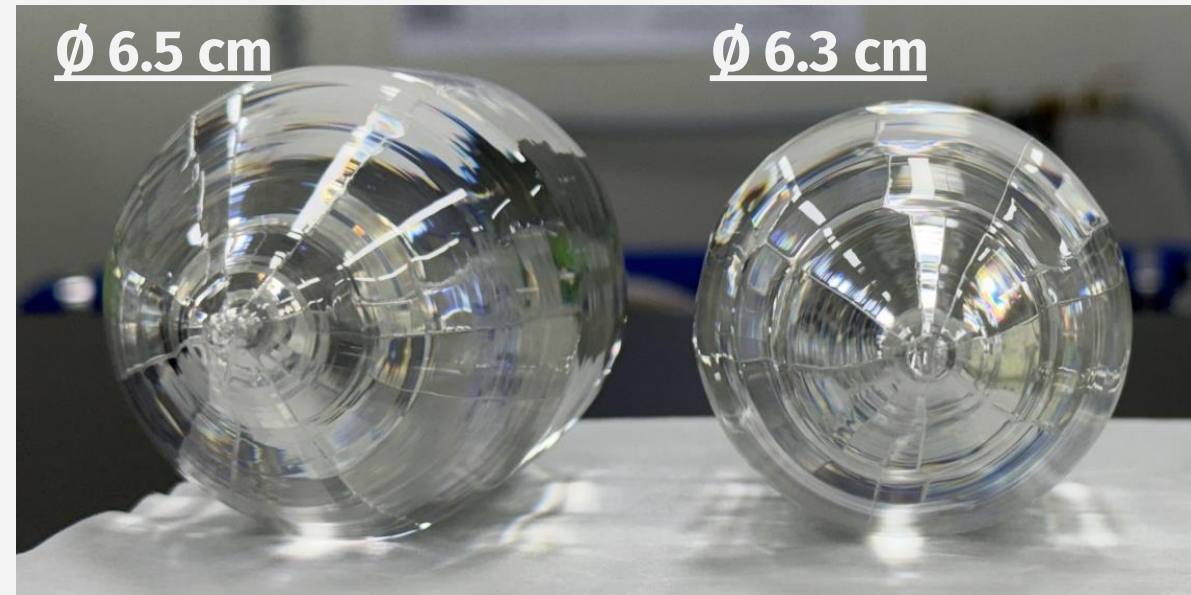
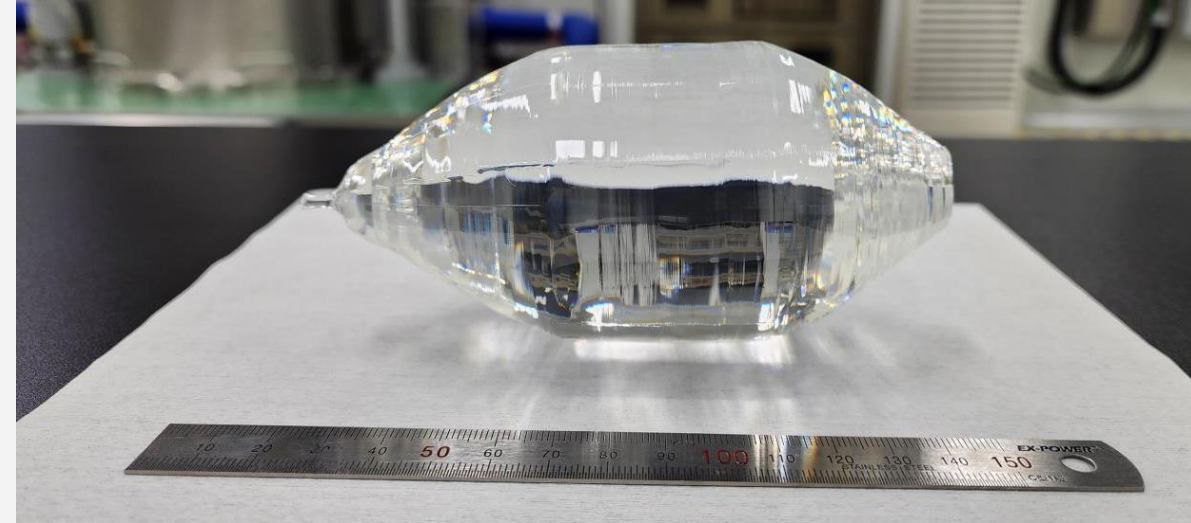
Element	Range of concentrations, [ppm]	Element	Range of concentrations, [ppb]
Al	0.5—2.1	Si*	$<1 \times 10^4$
K	0.3—1.6	W	10—1350
Fe	0.2—1.6	Sr	2—80
Ni	<0.02 —1.2	Ba	8—20
Cu	<0.1 —0.5	Pb	3—9
Cr*	<1	Th	0.03—0.15
Ti*	0.3—1.5	U	0.04—0.28



How LMO crystals grow at CUP?



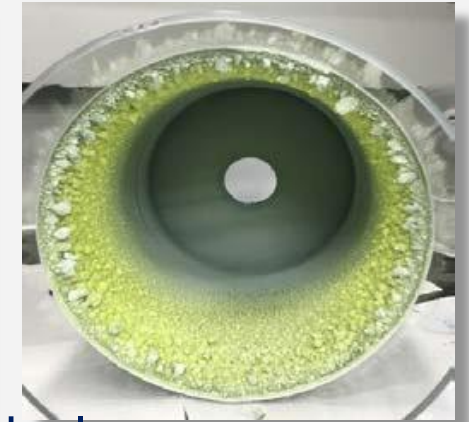
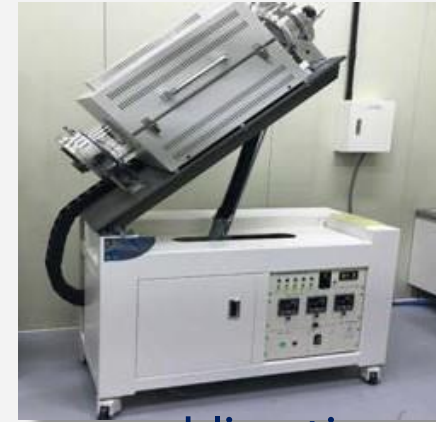
- **Three conventional Czochralski pullers** with Pt crucibles
- Advanced **50% crystallization** efficiency
- **High segregation factor** (up to five orders of magnitude) for radioimpurities.
- **Up to 9 crystals per month** is routine, but scalable.



Purification of $^{100}\text{MoO}_3$ for AMoRE-II detector

$^{100}\text{MoO}_3$ raw material 99.997% (>99.5% enrichment with Mo-100)

1. In-house designed sublimation apparatus: [JINST 14\(11\):T11002-T11002](#)
2. Wet chemistry: co-precipitation and membrane filtration. In-house purified co-precipitation agent.
3. Purification capacity at CUP: 5 kg/month
4. Recovery efficiency for the process 99%: [Frontiers in Physics 11](#)
5. Irrecoverable losses: 1%
6. Purity of the produced powder is controlled with ICP-MS and HPGe array at CUP.



1. sublimation under low vacuum



2. Wet chemistry



3. synthesis of ammonium polymolybdate powder and its annealing 4. final $^{100}\text{MoO}_3$ powder

ICP-MS assay of $^{100}\text{MoO}_3$ powder for AMoRE-II



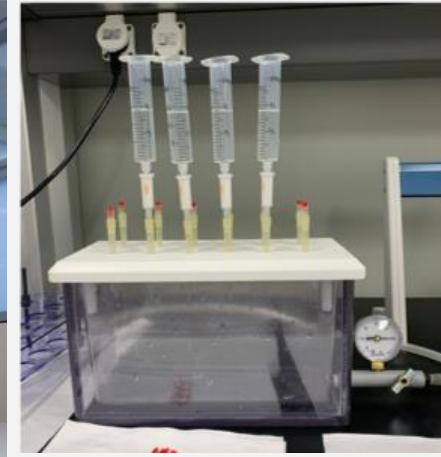
ICP-MS Agilent 7900



Class 1000 clean room



Microwave digestion



Extractive chromatography



Element	Al, [ppb]	K, [ppb]	W, [ppb]	Sr, [ppb]	Ba, [ppb]	Pb, [ppb]	Th, [ppt]	U, [ppt]
$^{100}\text{MoO}_3$ raw powders	500 - 2100	300 - 1600	10 - 1350	2 - 80	8 - 20	3 - 9	30 - 150	40 - 280
CUP PUR $^{100}\text{MoO}_3$ powders	<100 - 700	<500 - 1100	<50 - 700	<0.2	<4	<0.5	<7	<7
DF	1 - 20	1-3		10 - 400	2 - 5	6 - 18	3 - 20	5 - 40

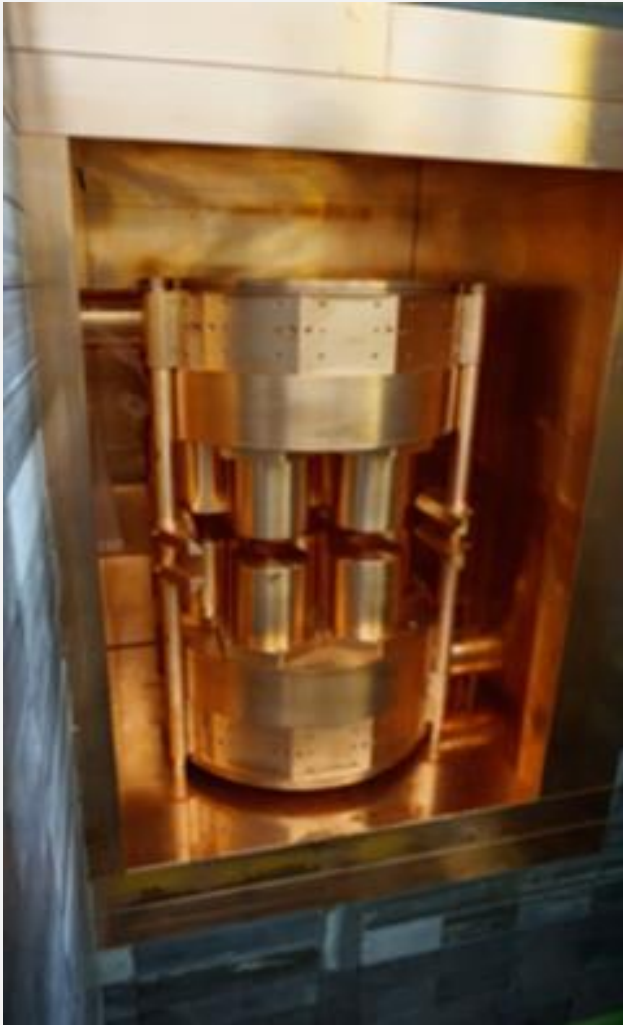
*DF stands for Decontamination Factor

HPGe assay of $^{100}\text{MoO}_3$ powder for AMoRE-II

HPGe array measurement of 13 kg of $^{100}\text{MoO}_3$ before and after purification at CUP

Activity, $\mu\text{Bq/kg}$	^{228}Ac	^{228}Th	^{226}Ra	^{40}K
$^{100}\text{MoO}_3$ raw powder	260 ± 50	210 ± 50	260 ± 50	8500 ± 1400
CUP PUR $^{100}\text{MoO}_3$ powders	<27	<16	110 ± 30	1700 ± 340
DF	9	13	2	5

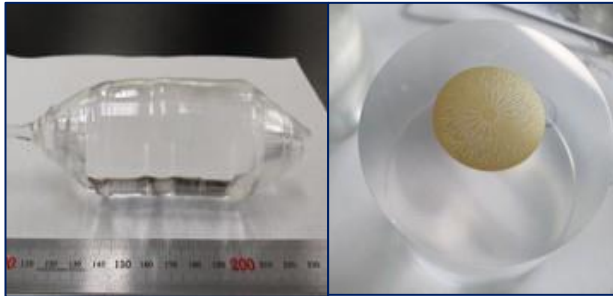
*DF stands for Decontamination Factor



The array of 14 HPGe detectors with inner copper shielding of ARRAY detector

Other candidates to be AMoRE detector

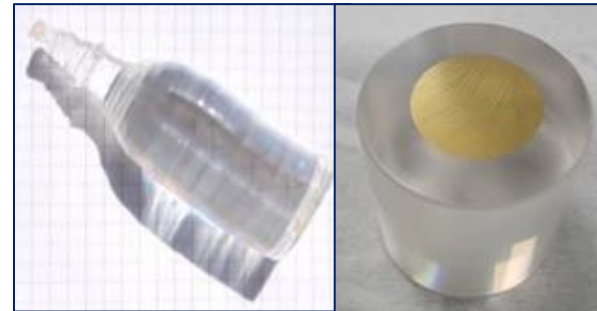
AMoRE-II crystal growing and purification R&D



Best radiopurity achieved:
[Frontiers of Physics 12\(2024\):1347162](#)
[Inorganics 11\(10\):410](#)



⁴⁸deplCa is required. Purification and recycling methods were developed at CUP:
[Journal of Material Cycles and Waste Management 21\(6\)](#)



No commercially available pure enough product:
[Inorganic Materials 56\(8\):867-874](#)



Archaeological Roman lead is required. Very difficult to recycle

Li₂CO₃ selection for AMoRE-II

5N purity does not guarantee the radiopurity

	⁴⁰ K	²⁰⁸ Tl	²²⁶ Ra (²¹⁴ Bi)	²²⁸ Ac
Alfa Aesar, Puratronic*, 99.998%				
2016	9 ± 3.4	0.41 ± 0.22	0.95 ± 0.22	1.4 ± 0.64
2021 1500 USD/kg	26.5 ± 7.1	<3.8	28 ± 2	<5.6
2017 [26]	≤42	n/d	705 ± 30	12 ± 4
NRMP powders				
TU 6-09-3728-83	<11.5	<1.8	<1.8	<1.7
Pharma grade	<66.4	9.1 ± 2.6	2,730 ± 137	108.2 ± 9.3
Tech. Grade LiOH	<5.1	<3.8	730 ± 20	28 ± 4
Li ₂ CO ₃ from Chinese market				
A, 5N	<16.6	<1.3	57.4 ± 3.2	6.3 ± 1.3
B, 5N	<10.9	11.4 ± 1.4	295 ± 15	12 ± 2
C, 5N	<86.7	185 ± 14	10,850 ± 540	435 ± 35

Unavailable
Yellowish
crystals

Unavailable
High Ra and
Th

Values and upper limits are given at 95% confidence interval. The “n/d” stands for no data.

Radioactivity reduction in Li_2CO_3 with CUP purification

Raw Li_2CO_3 , 5N

Radionuclides	Activity [mBq/kg]		Purified LiNO_3		Final Li_2CO_3
^{40}K	≤ 66		77 ± 6		≤ 14
^{137}Cs	≤ 3		1.2 ± 0.2		≤ 1.0
^{234}Th	≤ 151		≤ 16		≤ 22
$^{234\text{m}}\text{Pa}$	≤ 162		≤ 47		≤ 47
^{226}Ra	2730 ± 140		≤ 1.0		≤ 1.5
^{228}Ac	110 ± 10		≤ 1.4		≤ 1.4
^{228}Th	9 ± 3		≤ 1.0		≤ 1.4

We have developed several methods of purification to remove K, Th, U, and Ra contamination

- For mass-scale purification (over 10 kg per month), the construction of a special purification facility will be required

ICP-MS assay of each LMO crystal

Grower	Sample ID		K	Ba	Sr	Zr	Ir	Pb	Th	U
			(ppb)	(ppb)	ppt	ppt	ppt	ppt	ppt	ppt
C Z 0 1	CZ01-L2402ED-2	Top	<40	<3	<50	<100	<50	<200	<6	<6
		Bottom	<40	<3	<50	<100	<50	<200	<6	<6
	CZ01-L2403E-1	Top	<40	<3	<50	<100	<50	<200	<6	<6
		Bottom	<40	<3	<50	<100	<50	<200	<6	<6
	CZ01-L2404E-1	Top	<40	<3	<50	<100	<50	<200	<6	<6
		Bottom	<40	<3	<50	<100	<50	<200	<6	<6
	CZ01-L2501E-1	Top	<40	<3	<50	<100	<50	<200	<6	<6
		Bottom	<40	<3	<50	<100	<50	<200	<6	<6
C Z 0 2	CZ02-L2402ED-2	Top	<40	<3	<50	<100	<50	<200	<6	<6
		Bottom	<40	<3	<50	<100	<50	<200	<6	<6
	CZ02-L2403E-1	Top	<40	<3	<50	<100	<50	<200	<6	<6
		Bottom	<40	<3	<50	<100	<50	<200	<6	<6
	CZ02-L2404E-1	Top	<40	<3	<50	<100	<50	<200	<6	<6
		Bottom	<40	<3	<50	<100	<50	<200	<6	<6



- Each crystal is checked with ICP-MS, carefully polished and surface-cleaned, and stored in N₂ low-humidity box

COSINE-200 experiment

- The COSINE experiment searches for dark matter using ultra-low-background NaI(Tl) scintillating crystals to verify DAMA/LIBRA's claim with the same target material but improved radiopurity.



COSINE-100
(2016-2023)



COSINE-100U
upgrade



COSINE-200

Why ultra-pure NaI(Tl)?

- COSINE searches for dark matter using NaI(Tl) crystals
- Experimental sensitivity is limited by internal radioactive backgrounds
- Major contaminants: ^{40}K , ^{210}Pb

Goal

- Produce several hundred kg of ultra-pure NaI powder with $\text{K} < 20$ ppb and $\text{Pb} < 1$ ppb for the COSINE-200

Eur. Phys. J. C (2021) 81:837

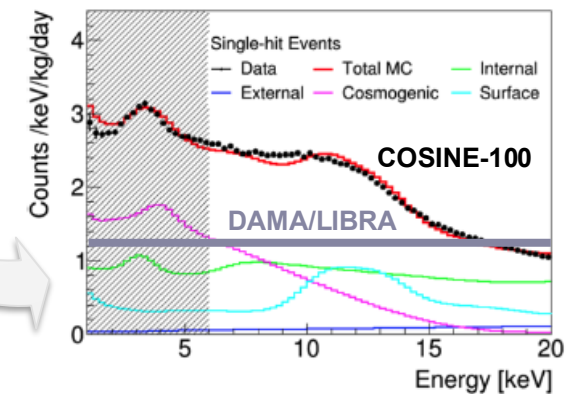


Fig. 9 [The low-energy spectra of single-hit events averaged for the five crystals. The measured energy spectrum after efficiency corrections [23] is compared with the total of the simulations. The range of 1–6 keV in the MC spectrum is extrapolated from the modeling]

Astropart. Phys. 141, 102709 (2022)

Large-scale NaI purification facility (initial charge up to 70 kg per batch)



(a) Full-scale facility



(b) feed tank



(c) mixing tank



(d) filtration unit



(e) receivers



(f) conical dryer

Adapted from Shin, K. et al., Front. Phys. 11, 1142849 (2023)

Radiopurity Evaluation by ICP-MS

COSINE-200 requirement

- K < 20 ppb
- Pb < 1 ppb

Sample No.	K (ppb)	Sr (ppb)	Ba (ppb)	Pb (ppb)	Th (ppt)	U (ppt)
Merck raw powder (Crystal grade), 2020	250±90	19±1	3.0±0.4	40±2	<6	<6
Merck raw powder (Crystal grade), 2021	730	42	146	58	<100	415
Merck raw powder (Astro grade), 2020	5±3	0.3±0.1	0.6±0.1	0.8±0.5	<6	< 6
Purified powder (23-1)	7	21	0.6	<0.3	<4.0	<4.0
Purified powder (22-5)	10.6	0.4	1.6	0.9	<100	<20
Residual Melt recovery powder (25-1)	7	0.2	0.9	<0.2	<4	<4
Residual Melt recovery powder (25-2)	7	0.2	0.7	<0.2	<4	<4
Mother solution recovery powder (22-2)	21	0.2	0.7	1	<4	<4



- ~400 kg of purified NaI powder with K < 20 ppb is currently stored
- Water content is monitored using Karl Fischer titration.
- Moisture level below 0.1% is required for stable crystal growth.
- Excessive moisture may damage the quartz crucible.

Nal(Tl) crystals grown using CUP-purified NaI powder

$^{210}\text{Pb} = <0.06 \text{ mBq/kg}$ for NaI-34 and NaI-35

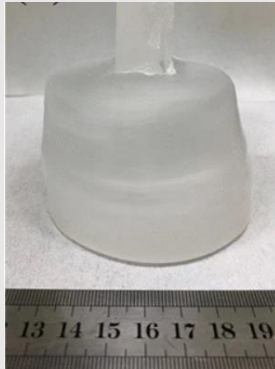
On pause now



Full size grower
for
~ 100 kg ingot

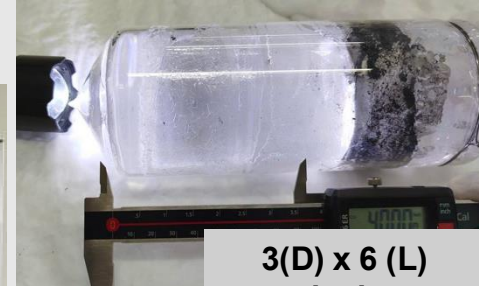


Test grower
~ 1kg ingot

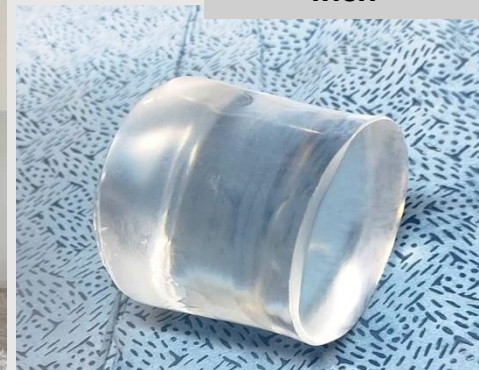


2.5 inch test
Kyropoulos growth
(CUP, Korea)

$^{210}\text{Pb} = 0.36(4) \text{ mBq/kg}$
 $^{\text{nat}}\text{K} \sim 8 \text{ ppb}$

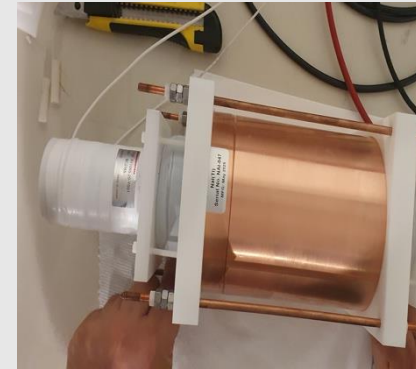


3(D) x 6 (L)
inch



3 inch
Bridgman growth
(KNU, Korea)

$^{210}\text{Pb} = 0.393(4) \text{ mBq/kg}$



Bridgman growth
(RMD, USA)

Images adapted from Lee, H. et al., Front. Phys. 11, 1142765 (2023) and Park, B. J. et al., Eur. Phys. J. C 80, 814 (2020).

Summary



- We have a full material purification, crystal growth, and surface treatment facility at CUP to get 360 AMoRE-II LMO crystals
- The facility could be scaled up for AMoRE-III or other large Mo-based crystal experiments
- NaI COSINE-200 powder has been proven as a reliable and, recently, the only available material for NaI-based experiments
- We are scaling up the Bridgman grower at CUP to grow NaI crystals up to 5 inches.
- However, we measured the lowest ^{210}Pb level with a Kyropoulos-grown crystal at CUP. We are looking for solutions to resolve thermal field issues in the large open crucible.